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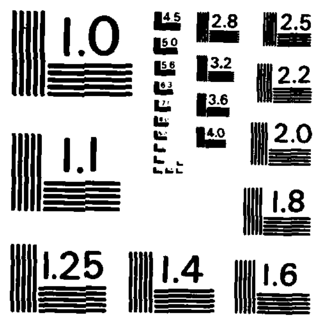

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Recently, Lai and Young (J. Acoust. Soc. Am., 72, 2000 (1982)) and Heritier (Opt. Communic. 44, 267 (1983)) have independently calculated the profile of a thermo-elastically generated opto-acoustic pulse due to the passage of an excitation laser pulse of certain spatial and temporal intensity distributions in a weakly absorbing medium. We have found that their results are essentially equivalent for Gaussian intensity distributipn of the excitation laser. We report the first experimental verification of such theoretical opto-acoustic profiles (over)

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excited by laser pulses of various pulse widths $\tau_{FWHM}$ (8nsec and 1.3 μsec) focussed to different beam waists $w_o$. Our observation of the profile is performed with a probe deflection technique instead of using conventional acoustic transducers which have limited risetimes and nonflat frequency response. Our results are in reasonable agreement with theory, and clearly indicate that sharp opto-acoustic pulses can be generated by laser pulses of short $\tau_{FWHM}$ and small $w_o$.

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# Profile of Laser-produced Acoustic Pulse in a Liquid

By

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# Research Report

## PROFILE OF LASER-PRODUCED ACOUSTIC PULSE IN A LIQUID

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# PROFILE OF LASER-PRODUCED ACOUSTIC PULSE IN A LIQUID*

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ABSTRACT: Recently, Lai and Young (*J. Acoust. Soc. Am.*, 72, 2000 (1982)) and Heritier (*Opt. Communic.* 44, 267 (1983)) have independently calculated the profile of a thermo-elastically generated opto-acoustic pulse due to the passage of an excitation laser pulse of certain spatial and temporal intensity distributions in a weakly absorbing medium. Lai's and Young's ~~We have found that their~~ results are essentially equivalent for Gaussian intensity distribution of the excitation laser. The authors ~~We~~ report the first experimental verification of such theoretical opto-acoustic profiles excited by laser pulses of various pulse widths tau $\tau_{FWHM}$ (8 nsec and 1.3 microsec ~~μsec)~~ focussed to different beam waists Sub 0 $w_0$. Our observation of the profile is performed with a probe beam deflection technique instead of using conventional acoustic transducers which have limited risetimes and nonflat frequency response. The authors' ~~Our~~ results are in reasonable agreement with theory, and clearly indicate that sharp opto-acoustic pulses can be generated by laser pulses of short $\tau_{FWHM}$ *and* small Sub 0 $w_0$.

*This work is partially supported by the Office of Naval Research.

## 1. INTRODUCTION

The phenomena of opto-acoustic (OA) pulse generation due to thermal expansion induced by a pulsed optical beam has been known for some twenty years now. Earlier observations of OA pulses produced by pulsed laser beams in liquids were reported, for example, by Askaryan *et al.*,[1] Carome *et al.*,[2] and Rentzepis and Pao.[3] Important modern applications of pulsed OA generation include spectroscopy,[4] energy transfer studies and ultrasonic measurements.[5] However, most experimental detection of the OA pulses have been performed by using piezoelectric transducers or microphones, and the true OA pulse profile produced by an excitation laser pulse cannot be readily measured. The present work seems to be the first quantitative experimental investigation of the true OA profile (detected by a probe beam deflection technique[6,7,8]) as a function of the excitation laser pulse parameters. Such quantitative understanding is important for the optimization of various OA applications.[5,9]

The importance of theoretical understanding of the pulsed thermo-elastic OA generation phenomenon can be appreciated from the many theoretical papers on this topic, including White,[10] Gournay,[11] Hu,[12] Lyamshev and Naugol'nykh[13] and others. Recently, explicit mathematical forms for the OA pulse profile in a liquid excited by a Gaussian (spatial and temporal) laser pulse have been given independently by Lai and Young[14] and Heritier.[15] Both theoretical treatments have provided insight as to the method to produce OA pulses of desired profile; for example, laser pulses of narrow diameter and short duration should produce narrow OA pulses propagating in the lateral direction in a weakly absorbing medium. Their theoretical results, which turn out to be basically equivalent, are described in Section 2. Our experimental arrangement to verify these theoretical thermo-elastic OA profiles are described in Section 3, and our observations are presented in Section 4.

## 2. THEORY

Analytical solution of the pulsed OA generation due to thermo-elastic mechanism in a weakly absorbing medium is possible for the simple case of a Gaussian intensity distribution in space and time of the excitation laser beam. It should be noted that the exact form for the Gaussian distribution may vary among authors. Lai and Young[14] have described a Gaussian laser beam in the following form:

$$I_{LY}(r,t) = \frac{E}{(2\pi)^{3/2}R^2\tau_p} \exp\left(-\frac{r^2}{2R^2} - \frac{t^2}{2\tau_p^2}\right) \tag{1}$$

where $I_{LY}$ is the laser intensity at distance r from the beam axis, E is the laser pulse energy, t is the time from the peak of the laser beam, R is a measure of the laser beam radius and $\tau_p$ is a measure of the pulse duration. Lai and Young have solved the acoustic wave equation with a source term being due to the thermal expansion produced by $I_{LY}(r,t)$, and obtained the OA pressure $P_{LY}(r,t)$ in terms of Bessel function:

$$P_{LY}(r,t) = K\tau_e^{-3/2} \frac{d\Phi_o(\xi)}{d\xi} \tag{2}$$

where

$$K = \frac{\alpha\beta E}{8\pi^{1/2}C_p} \left(\frac{v_a}{r}\right)^{1/2}, \tag{3}$$

$$\tau_e = [\tau_p^2 + \tau_a^2]^{1/2}, \tag{4}$$

$$\Phi_o(\xi) = |\xi|^{1/2} \exp(-\xi^2/4)\left[\frac{\sqrt{2}}{\pi}K_{1/4}\left(\frac{\xi^2}{4}\right) + 2\Theta(\xi)I_{1/4}\left(\frac{\xi^2}{4}\right)\right], \tag{5}$$

$$\xi = \left(t - \frac{r}{v_a}\right)/\tau_e = t'/\tau_e. \tag{6}$$

In Eq. (2), K determines the magnitude of the OA pulse, $\tau_e$ is a time scale factor which determines the width and the magnitude of the OA pulse, and $d\Phi_o/d\xi$ determines the shape of the pulse. The symbols used have the following meanings: $\alpha$ is the optical absorption coefficient, $\beta$ is the volume expansivity, $C_p$ is the specific heat at constant pressure, $v_a$ is the acoustic velocity, $\tau_a = R/v_a$ is an acoustic transit time in the source, $t'$ is the retarded time with $t'=0$ being at the moment $r/v_a$ after the laser peak, $\xi$ is a normalized retarded time, $K_{1/4}$ and $I_{1/4}$ are Bessel functions of imaginary arguments and $\Theta$ is the unit step function.

Independent of Lai and Young,[14] Heritier[15] has also solved the thermo-elastic wave equation for excitation by a Gaussian laser beam, with an intensity distribution $I_H(r,t)$ defined by

$$I_H(r,t) = \frac{2E}{\pi^{3/2} w_o^2 \tau} \exp\left( - \frac{2r^2}{w_o^2} - \frac{t^2}{\tau^2} \right) . \tag{7}$$

Here $w_o$ is known as the beam waist [16] and is equal to 2R, and $\tau$ is the $e^{-1}$ half width of the laser pulse, related to the full width at half maximum $\tau_{FWHM}$ by

$$\tau_{FWHM} = 1.665\ \tau . \tag{8}$$

The solution for the OA pressure $P_H(r,t)$ obtained by Heritier due to the thermal expansion produced by $I_H(r,t)$ in a weakly absorbing medium is

$$P_H(r,t) = K' \epsilon^{-3/2} F(x) \tag{9}$$

where

$$K' = \frac{\alpha\beta E}{(2\pi)^{3/2} C_p} \left( \frac{v_a}{r} \right)^{1/2} , \tag{10}$$

$$\varepsilon = (\tau^2 + w_0^2/2v_a^2)^{1/2} \tag{11}$$

$$x = \left(t - \frac{r}{v_a}\right)/\varepsilon = t'/\varepsilon \tag{12}$$

and

$$F(x) = \left[\Gamma\left(\frac{3}{4}\right){}_1F_1\left(-\frac{1}{4};\frac{1}{2};x^2\right)\right.$$

$$\left. - 2x\Gamma\left(\frac{5}{4}\right){}_1F_1\left(\frac{1}{4};\frac{3}{2};x^2\right)\right] \exp(-x^2)\ . \tag{13}$$

Here, analogous to the Lai and Young's solution in Eqs. (2)-(6), $K'$ is an amplitude factor, $\varepsilon$ is a time scale factor and F is the shape function expressed in terms of confluent (or degenerate) hypergeometric function ${}_1F_1(a;b;z)$. The symbols not previously introduced are x which is the retarded dimensionless time equal to the retarded time $t'$ divided by $\varepsilon$, and $\Gamma$ which is the Gamma function.

We have made comparison of the numerical resuits of Lai and Young's solution (Eqs. (2-6)) and those of Heritier's solution (Eqs. (9-13)). Both solutions state that the shape of the OA pressure pulse is dependent on time t only through the retarded dimensionless time $\xi = t'/\tau_e$ in Lai and Young's result and $x = t'/\varepsilon$ in Heritier's result. Due to their different definitions of the Gaussian spatial and temporal dependence of the laser pulse, it should be noted that the two "time scale parameters" $\varepsilon$ and $\tau_e$ differ by a factor of $\sqrt{2}$:

$$\varepsilon = \sqrt{2}\,\tau_e\ . \tag{14}$$

We have found that the solution for the time dependent OA pressure profiles $P_{LY}(r,t)$ and $P_H(r,t)$ given in Eqs. (2) and (9), respectively, are essentially equivalent; this is shown in

Fig. 1, where both theoretical profiles are plotted with respect to the dimensionless times $\xi$ and x.

To be specific, we shall use Heritier's[15] notation from here on. $\varepsilon$ is called the "time scale parameter" of the OA pulse, because $\varepsilon$ truly controls the temporal width of the OA pulse. For example, Heritier's[15] line shape (Fig. 1) shows that the separation between the peak and the trough of the OA pressure pulse is $\Delta x = 1.66$, corresponding to a time separation being:

$$\Delta t_{pk\text{-}tr} = 1.66\varepsilon \ . \tag{15}$$

The same numerical result is obtained by analyzing Lai and Young's results.

## 3. EXPERIMENT

The present experiment is a first attempt to verify the theoretical OA pulse profile predicted by Lai and Young[14] and Heritier.[15] The schematics of our experimental arrangement is shown in Fig. 2. Two types of excitation lasers with different pulse durations are used. Most data were obtained with a Molectron $N_2$ pumped dye laser providing laser pulses of $\tau_{FWHM}=8$ nsec, and pulse energy E=0.16 mJ at 10 Hz repetition rate. The dye laser wavelength is 580 nm with a bandwidth of 5 nm. The laser beam is spatially filtered and an iris is used to provide a 4 mm diameter collimated beam, which is directed to a 90° prism and a focussing lens mounted on a translation stage (see Fig. 2). This excitation laser pulse is thus focussed into the sample cell. Some of our data have also been obtained[6] with an excitation laser of longer duration ($\tau_{FWHM}=1.3$ $\mu$sec). To change the beam waist $w_o$ of the excitation laser, we use focussing lenses of different focal lengths f. For the $N_2$ pumped dye laser, values for f used are 60 mm, 120 mm and 254 mm; the corresponding values of

$w_o$ are measured by using a calibrated pinhole to be 20, 49 and 74 $\mu m$ respectively. For the flashlamp pumped dye laser, f is 80 mm, and the waist $w_o$ is approximately 10 $\mu m$.

The probe laser beam from a 1 mW HeNe laser is beam expanded to a 10 mm diameter collimated beam, which is focussed in the cell by a lens of 135 mm focal length, producing a probe beam waist of about 9 $\mu m$. The direction of the probe beam is adjusted to be parallel to that of the excitation beam. This is done by first moving the translation stage controlling the excitation laser beam so that the two beams are made to be coincident. Subsequently, the two beams are displaced (by moving the translation stage) to a separation of typically 5 mm for the present measurements. The glass sample cell is of rectangular shape ($5 \times 3 \times 8$ cm) and contains methanol doped with a dye (*e.g*, Rhodamine 640) at about 23°C. The observed OA signal shape is independent of the dye concentration if the transmission of each laser beam is not less than ~30%.

The transmitted probe beam is directed onto a 6328Å interference filter (to block out scattered light from the dye laser) and a fast photodiode-preamplifier assembly (Analog Module model LNVA-O). The small active area of the photodiode (0.8 $mm^2$) measures the intensity of only a small portion of the probe beam; its position in the probe beam (of diameter about 4 mm at the photodiode) can be adjusted by a translation stage. When the OA pulse produced by the excitation beam crosses the probe beam, a transient angular deflection[17] $\phi$ of the probe is produced:

$$\phi(r,t) \approx \frac{\ell}{n_o} \frac{\partial n(r,t)}{\partial r} \propto \frac{\partial p(r,t)}{\partial t} \tag{17}$$

where $n_o$ is the normal refractive index of the liquid, n(r,t) is the change in the refractive index which is proportional to the OA pressure P(r,t), and $\ell$ is the interaction length of the OA pulse with the probe laser. The small transient probe deflection $\phi$ causes the probe

beam to move across the detection photodiode. If the photodiode is suitably positioned at a wing of the probe beam cross section, the deflection $\phi$ produces an optimum intensity change; however, if it is positioned at the center of the probe beam, $\phi$ produces much smaller intensity changes. This is explained in Fig. 3, where the observed signals are also shown. The photodiode and preamp assembly has a bandwidth of 100 Hz-100 MHz. Its output is displayed on a 75 MHz bandwidth oscilloscope (Tektronix 7854 with 7A24 plug in). The digitizing and signal averaging capability of the oscilloscope is too slow to be useful for recording the OA signal; hence, any signal averaging is presently performed by photographing the scope display with multiple exposures (*e.g.*, for 50 laser shots).

The observed probe-beam deflection signal S(t) from the photo-diode is given by the following equation:

$$S(t) = G I_p'(r_d) L \phi \;, \tag{18}$$

where G is a constant depending on the photo-diode sensitivity and electronic gain of the detection system, $I_p'(r_d)$ is the lateral spatial derivative of the probe-beam intensity distribution at the photo-diode position $r_d$ (with the active area of the photo-diode being sufficiently small), and L is the "lever arm" of the probe beam (*i.e.*, distance from the interaction region in the cell to the photo-diode). Combining Eq. (17) and Eq. (18), we have

$$S(t) \propto \partial p(r,t)/\partial t \;, \tag{19}$$

which means that our experimental probe-beam deflection signal is a measure of the *time-derivative* of the OA pulse at the probe-beam position in the liquid. To compare with theory, we should either compare the observed signal with the time-derivative of the theoretical OA profile, or equivalently, compare the time-integral of the observed signal with

the theoretical OA profile. We shall choose the former method, since we believe that it is better to make theoretical comparisons with the raw data rather than with "processed" data wherever possible.

## 4. EXPERIMENTAL RESULTS

The experimentally observed probe deflection signals for three different focussing of the $N_2$ pumped dye laser are shown in Fig. 4. These signals are obtained with the photodiode being in a wing of the probe beam cross section (Fig. 3a) so that the signals are maximized. These observed signals are replotted in Fig. 5 in dimensionless times; the best fits to the theoretical deflection signal are obtained with the time scale parameter $\varepsilon$ being 10.6, 30.1 and 42.9 nsec for focussing lens of focal length 60, 120 and 254 mm, respectively. Our results are summarized in Table I, which gives the measured beam waist $w_o$ and the corresponding theoretical values of $\varepsilon$ according to Eq. (11), and the best fit values of $\varepsilon$. We see that the theoretical time scale parameter according to Eq. (11) agrees with the value obtained by fitting the experimental profile to better than about $\pm 20\%$. This provides a first experimental verification of the theoretical profiles calculated.[14,15]

The observed probe deflection signal for the case of flashlamp pumped dye laser excitation is shown in Fig. 6; the best fit to the theoretical signal (Fig. 7) is obtained with $\varepsilon = 1.05$ $\mu$sec. Here, if we fit the excitation laser pulse of $\tau_{FWHM} = 1.3$ $\mu$sec (see Fig. 6) by a Gaussian intensity distribution, we obtain a laser width parameter $\tau = 780$ nsec. For such a long pulse duration and small beam waist ($w_o = 10$ $\mu$m), Eq. (11) indicates that $\varepsilon$ should be approximately equal to $\tau$. The ~25% difference between the fitted OA time scale parameter $\varepsilon$ and laser pulse width $\tau$ is partially because the laser intensity is significantly nonGaussian.

It is clear from Figs. 5 and 7 that there is an imperfect agreement between the experimentally observed OA signal and the theoretical signal predicted by Lai and Young[14] or by Heritier.[15] The agreement is better for the earlier half of the signal than the latter half. The time scale parameter $\varepsilon$ obtained by fitting the observed signal to the theoretical one agrees with the value calculated from Eq. (11) to better than ±30%. Some of the reasons for the limited agreement are the following: (a) Neither the spatial nor the temporal dependence of the excitation laser pulse is exactly Gaussian, as the theoretical results assumed; (b) The excitation and probe laser beams may not be exactly parallel, and they are also not of infinite lengths with uniform cross sections, so that the theoretical one-dimension treatment is not totally valid.

## 5. CONCLUSION

We have used a probe beam deflection technique to measure the time-derivative of the pulsed OA signal in a liquid generated by an excitation laser pulse of $\tau_{FWHM}$ being 8 nsec or 1.3 $\mu$sec. The observed signals are in reasonable agreement with the theoretical results of Lai and Young[14] and Heritier,[15] which we have shown numerically to be equivalent.[18] We have thus experimentally confirmed (to an accuracy of ±30%) that the width of the pulsed OA signal is controlled by the magnitude of $\tau_e$, as given by Eq. (4), or equivalently by the magnitude of $\varepsilon$, as given by Eq. (11). Thus, sharp OA pulse is generated by a narrow laser pulse of short duration $\tau$ *and* focussed to a small beam waist $w_o$. For example, commerical lasers are readily available with $\tau \approx 0.5$ nsec; if the laser beam is focussed to a waist $w_o$ of 1 $\mu$m, this would produce an OA signal with a time scale parameter $\varepsilon \approx 0.8$ nsec. Since this corresponds to a peak to trough time separation of 1.33 nsec (see Eq. 15), a very sharp acoustic pulse with Fourier frequencies into the GHz regime is produced in the sample. The monitoring of the propagation of such an acoustic pulse in the sample should provide a novel

method of frequency multiplexed measurement of ultrasonic dispersion and attenuations in matter.[9]

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**TABLE I**

**Summary of Experimental Parameters for Our Study of OA Pulse Generation by a $N_2$ Pumped Dye Laser Beam ($\tau$=4.8 nsec).**

| Focal length f | Measured beam waist $w_o$ | $\varepsilon$ according to Eq (11) with measured $w_o$ and $\tau$=4.8 nsec | $\varepsilon$ obtained by a best theoretical fit of the observed probe deflection signal |
|---|---|---|---|
| 60 (mm) | 20 ($\mu$m) | 13.6 (nsec) | 10.6 (nsec) |
| 120 | 49 | 32.3 | 30.1 |
| 254 | 74 | 47.6 | 42.9 |

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18. We have been informed in private communication with K. Young of the Chinese University of Hong Kong that it is possible to show directly the mathematical equivalence of the theoretical results of Lai and Young[14] and Heritier.[15]

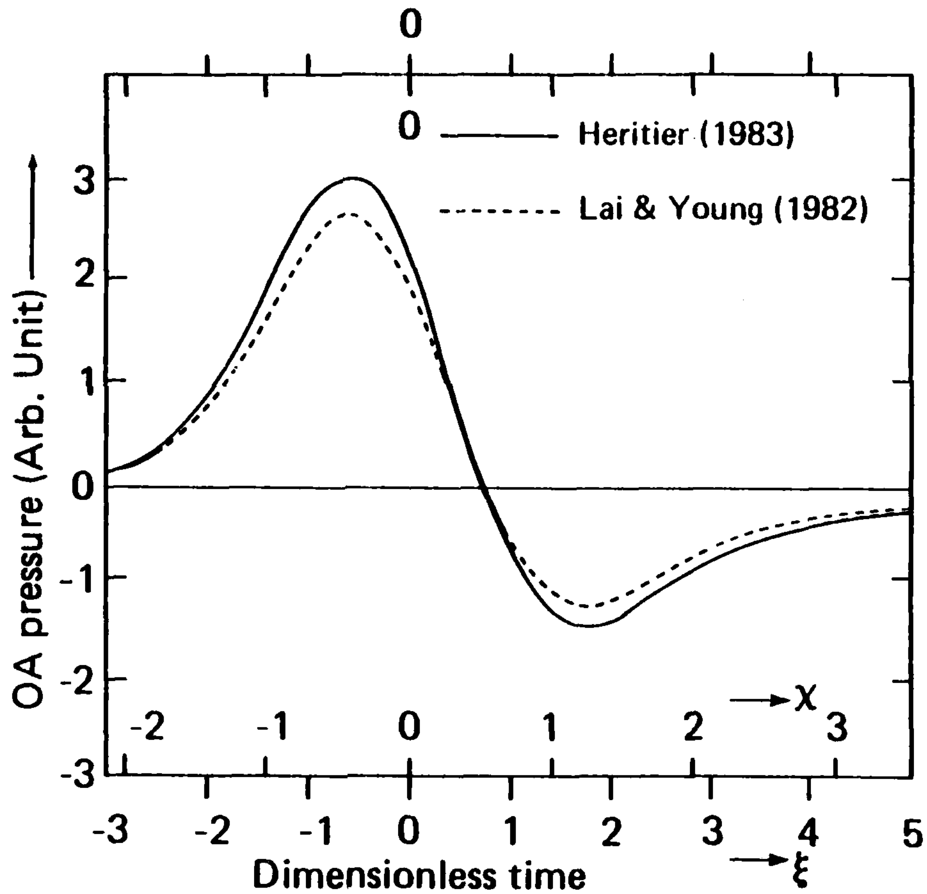


Figure 1. Theoretical OA pressure profiles according to Lai and Young[14] and to Heritier.[15] The amplitudes of the two profiles are made slightly different to show clearly the shape of each.

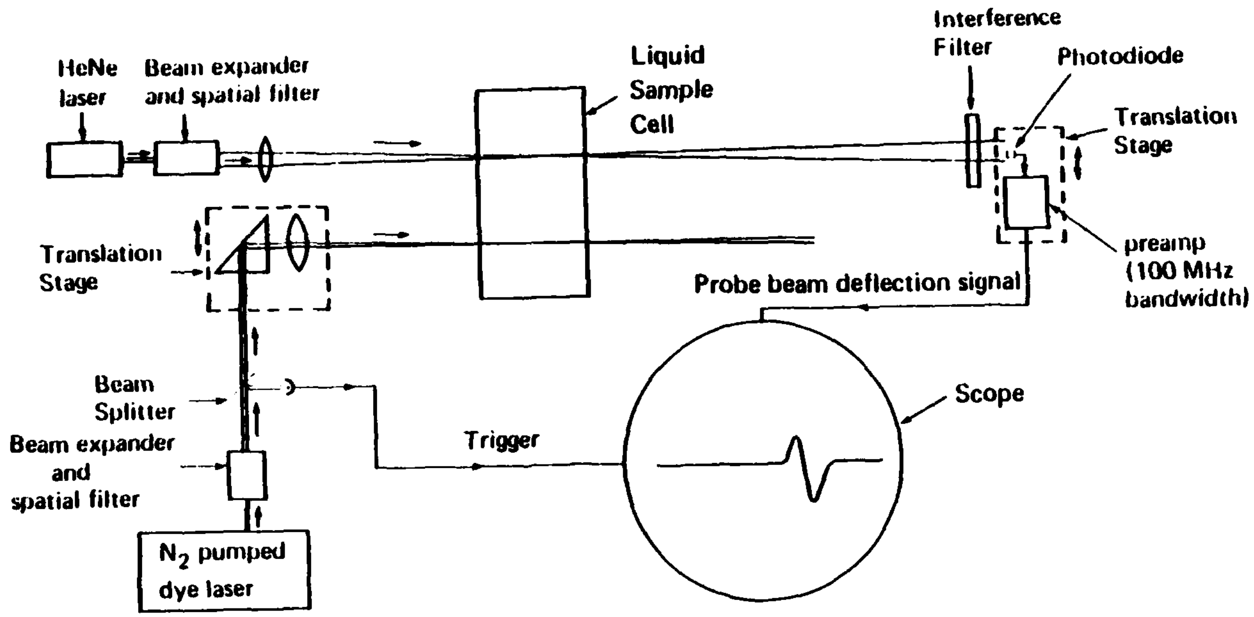


Figure 2. Experimental arrangement to measure the pulsed OA profile in liquids by a probe beam deflection technique.

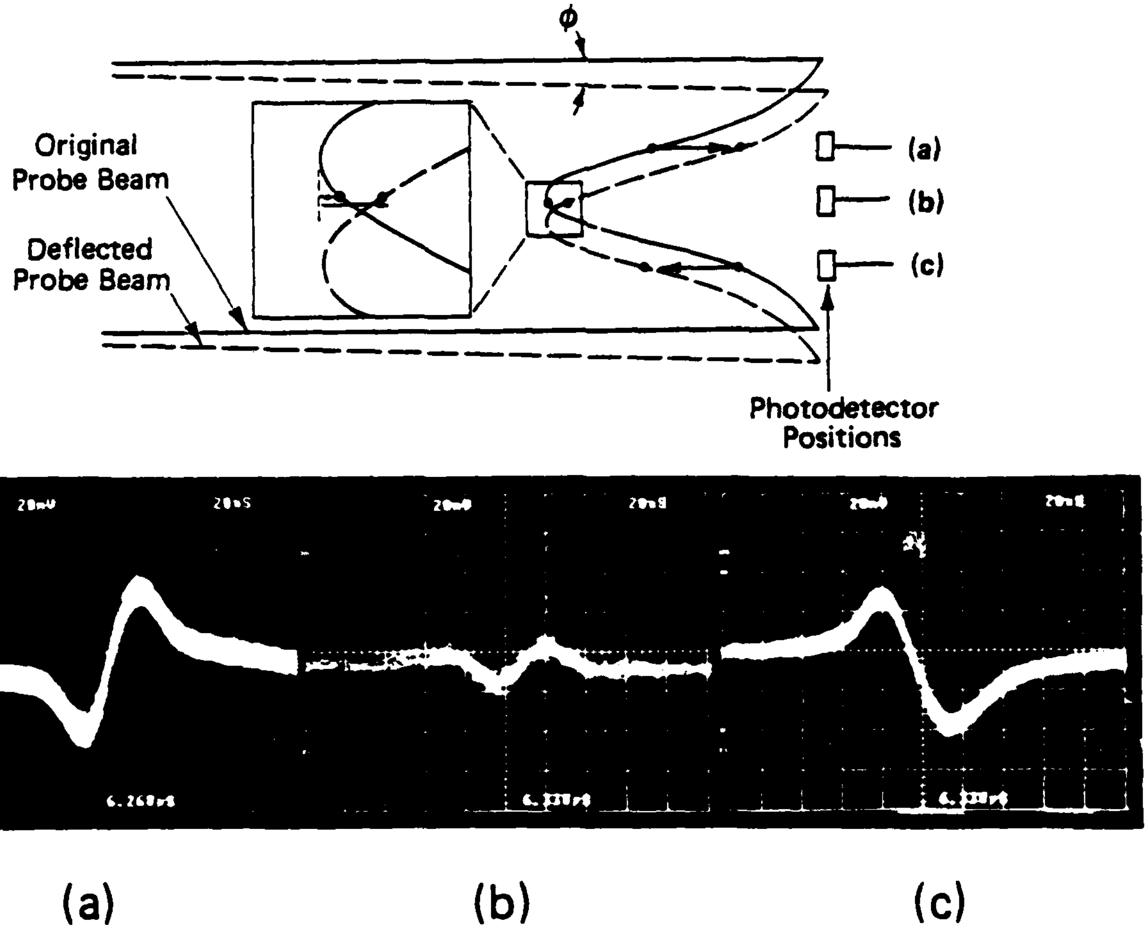


Figure 3. Observed probe beam deflection signal depends on the position of the small photodiode in the probe beam cross section. This is shown in the observed signals shown in the scope photos for the photodiode being on one wing, on center and on the other wing of the probe beam cross section, respectively. The inset explains how a deflection $\phi$ of the probe beam produces different photodiode signals for the different cases.

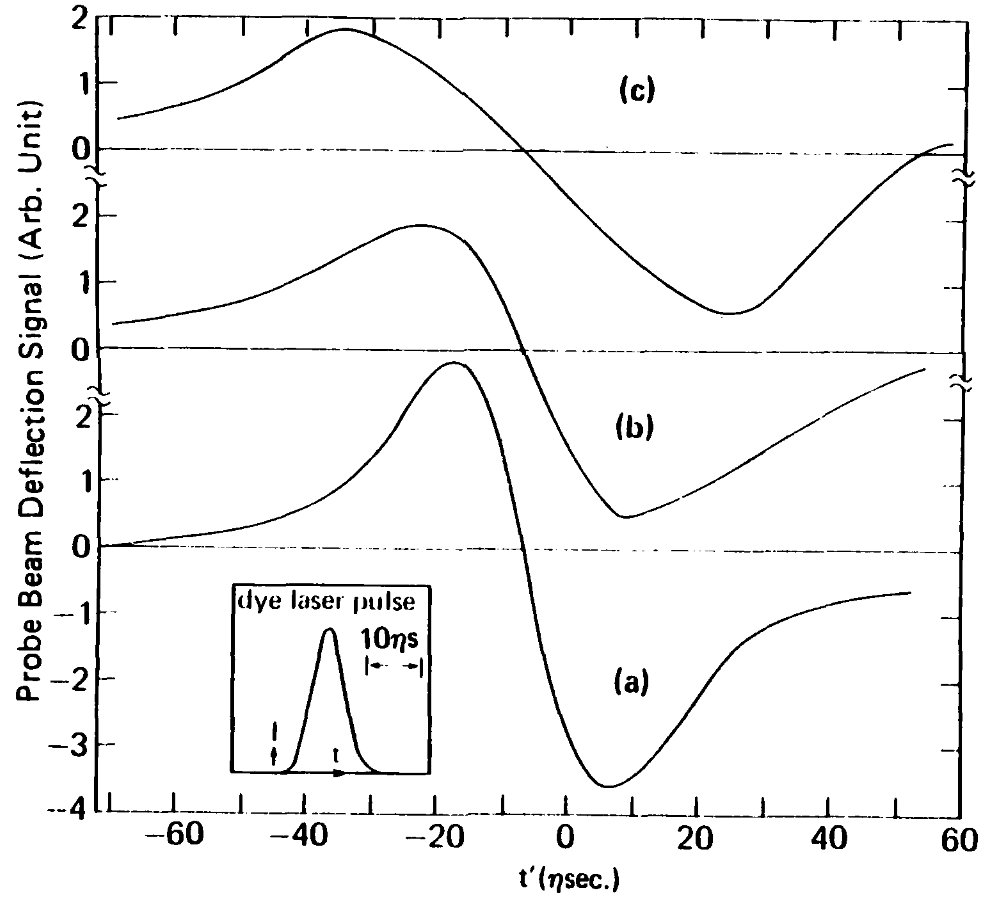


Figure 4. Observed probe beam deflection signals excited by various focussing of the $N_2$ pumped dye laser. The focal lengths f used are (a) 60 mm, (b) 120 mm and (c) 254 mm. The inset shows the temporal profile of the excitation laser pulse.

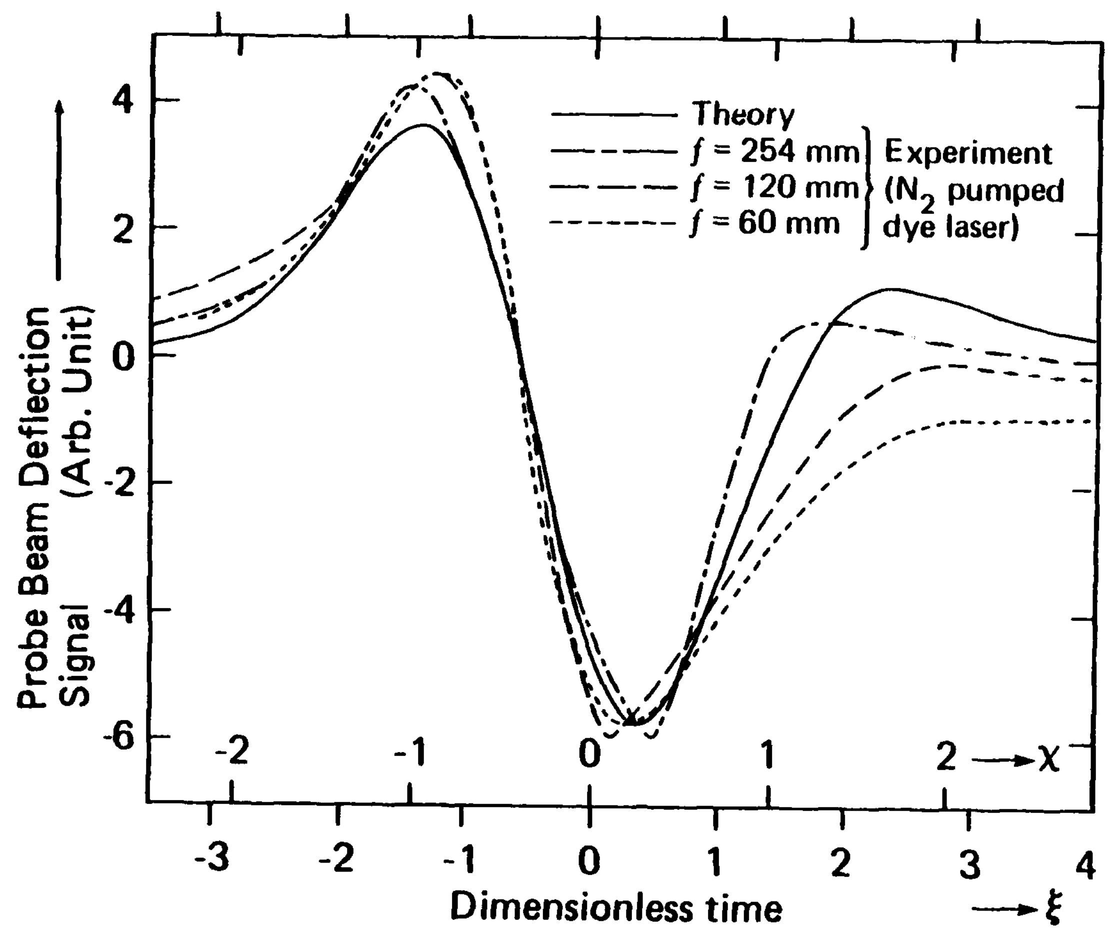


Figure 5. Comparison of theoretical probe beam deflection signal with the observed signals of Fig. 4 plotted against the dimensionless time x, with $\varepsilon$ used being 10.6, 30.1 and 42.9 nsec for (a), (b) and (c), respectively.

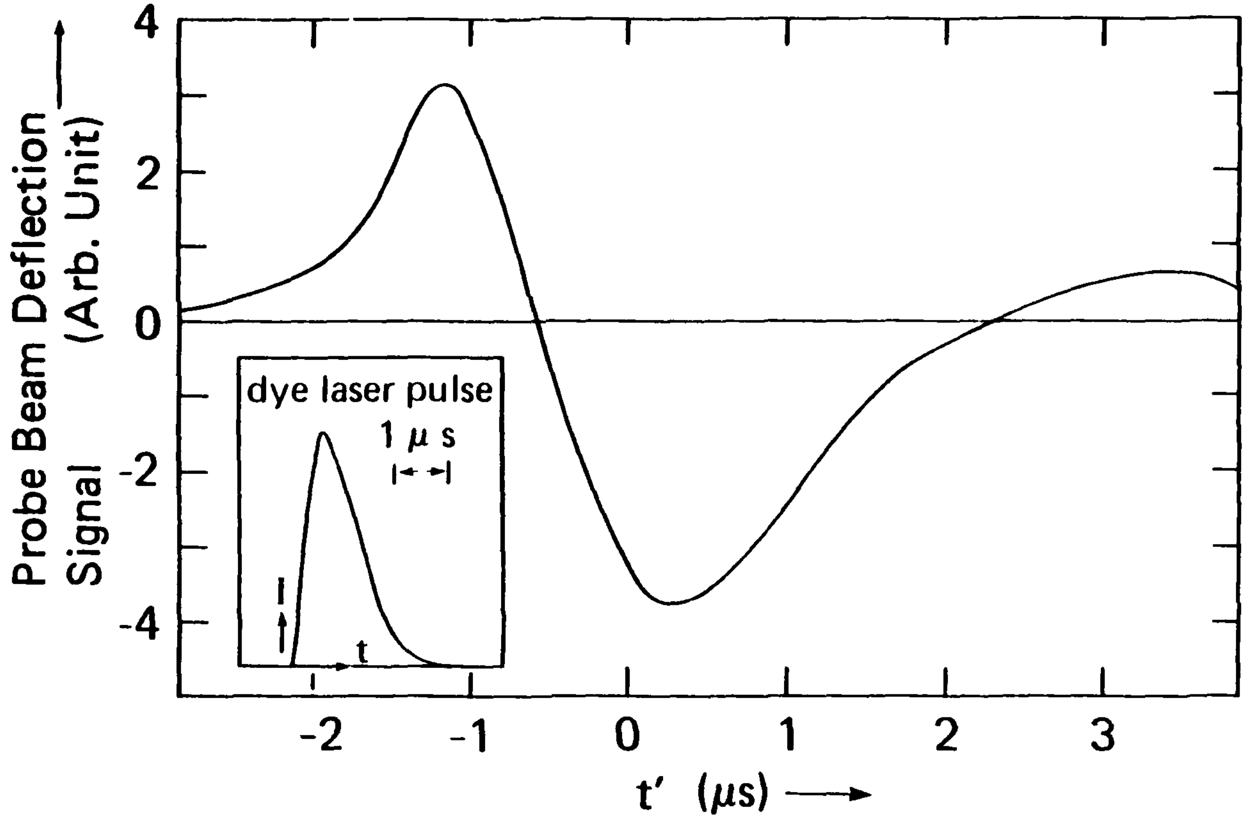


Figure 6. Observed probe beam deflection signal for excitation by the flashlamp pumped dye laser. The inset shows the temporal profile of the excitation laser pulse.

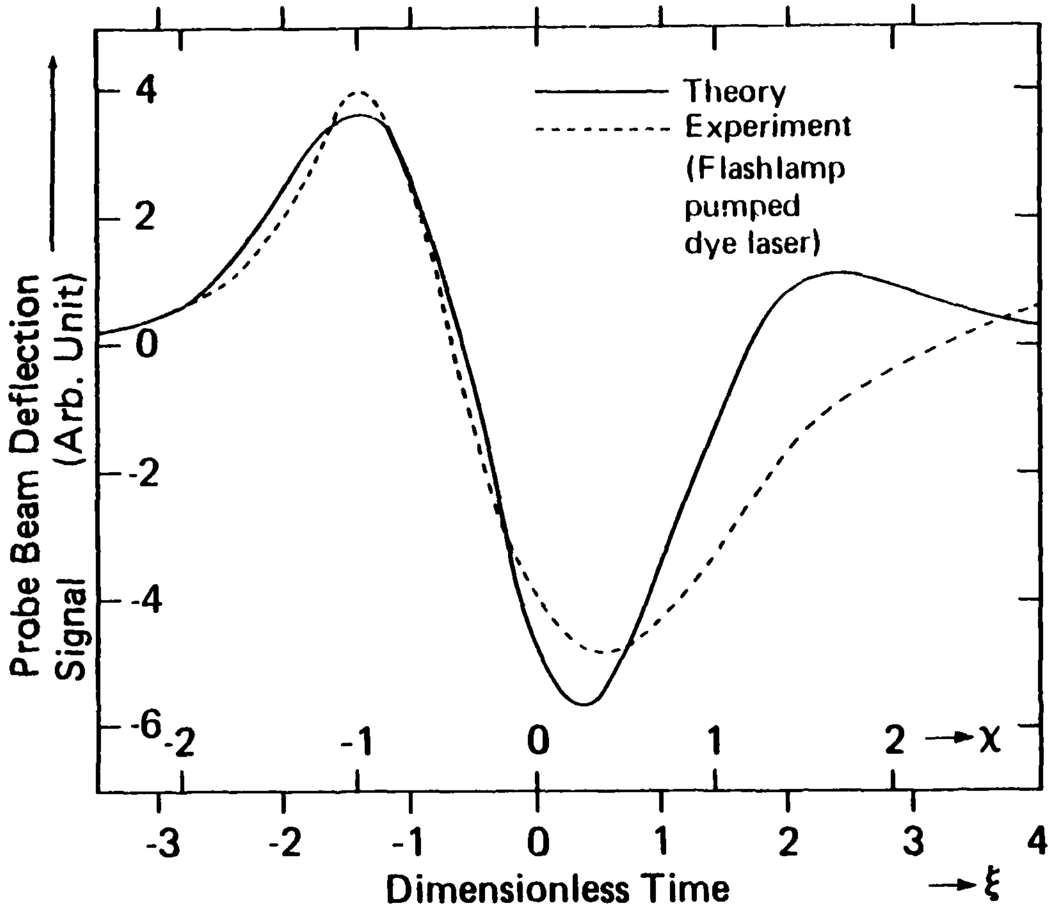


Figure 7. Comparison of theoretical probe beam deflection signal with the observed signal of Fig. 6 plotted against the dimensionless time x, with $\epsilon$ used being 1.05 $\mu$sec.